

# Synthesis, Characterization and Antimicrobial Activity of Some Organotin (IV) Complexes with a Potassium Hydrogen Ethanedioate Ligand

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**Abstract:** Four series of organotin (IV) derivatives of potassium hydrogen ethanedioate:  $Bu_2SnL_2$ ,  $Bu_3SnL$ ,  $Ph_2SnL_2$  and  $Ph_3SnL$  have been synthesized characterized by tin content analysis, FTIR,  $^1H$  NMR and  $^{13}C$  NMR and tested for antimicrobial activity against four Gram-positive bacteria: *Staphylococcus aureus*, *Streptococcus pyogenes*, *Bacillus cereus*, *Corynebacterium ulcerans*, four Gram-negative Bacteria: *Escherichia coli*, *Klebsiella pneumoniae*, *Proteus mirabilis* and *Pseudomonas aeruginosa* and four fungi strains of *Microsporium spp*: *Microsporium audouinii*, *Microsporium distortum*, *Microsporium gallinae* and *Microsporium gypseum*. Result showed that the compounds synthesized in general exhibited greater antimicrobial activity than potassium hydrogen ethanedioate. Organotin moieties play a vital role in deciding the antimicrobial activity of an organotin compound, this is true in this work. The order for the antifungal activity in this study was  $Bu_3SnL > Ph_3SnL > Ph_2SnL_2 > Bu_2SnL_2$ . The order of activity against gram-positive bacteria was  $Ph_3SnL > Bu_2SnL_2 > Ph_2SnL_2 > Bu_3SnL$ . The order of activity against gram-negative bacteria was  $Bu_3SnL > Bu_2SnL_2 > Ph_2SnL_2 > Ph_3SnL$ .

**Keywords:** Organotin derivatives, potassium hydrogen ethanedioate, Spectroscopic techniques, Antimicrobial activity

## 1. Introduction

Organotin complexes are known to attract more attention in recent years owing partly to their potential pharmaceutical value [1] as well as their potentials as biocides, homogenous catalysts, antifouling agents [2], antineoplastic [3] and anti-tuberculosis agents [4]. As a result, they have found wide application in agriculture, industries and medicine [5]. The high coordination ability of tin, especially its ability to participate either in weak or strong intra or intermolecular coordination [6], has given organotin (IV) chemistry wide recognition. Tin compounds have a fascinating solution and solid phase chemistry [7]. The coordination characteristics of tin and the structures differ dramatically in solution and in solid state when a nucleophilic group is present in the tin moiety or when an organic group is linked by a Sn—C bond [8] and bears additional functionality with donor properties due to the presence of O, N and S as donor atoms [2]-[6].

Organotin (IV) carboxylates are widely studied compounds with structural diversity and finds pharmaceutical applications [2] especially with reference to their antitumour [8] and anti-tuberculosis [9] activities. In the presence of additional coordinating atoms, some organotin carboxylates have fascinating structures, such as hexameric cyclic forms with a wide variety of coordination geometries [4], [10]. The combination of steric and electronic factors determine the specific structure adapted by a particular carboxylate [4], which could be monomeric, dimeric, tetrameric, oligomeric, cyclic and hexameric drum. It has been reported that the different structural types are formed due to the presence of additional coordinating sites (S, N or O) along with a carboxylic moiety [7]. In recent years, organotin (IV) carboxylates have been a subject of interest for some time because of their biochemical and commercial applications.

Generally, the biochemical activity of organotin (IV) carboxylates is greatly influenced by the structure of the molecule and the coordination number of the tin atom. Recognition of the importance between the biological properties and the structure of organotin (IV) carboxylates has stimulated the study of carboxylates of tin [11].

In an attempt to further explore the interesting features of organotin compounds based on the above, we report here the synthesis, characterization and antimicrobial properties of four organotin (IV) dicarboxylates prepared from four parent organotin compounds and ethanedioic acid.

## 2. Materials and Methods

Glass wares and Dean-Stack apparatus with standard quick fit joints were used throughout the work after cleaning and drying at 120 °C. Ethanedioic acid, KOH, parent organotins: dibutyltin (IV) oxide:  $(C_4H_9)_2SnO$ , tributyltin (IV) hydroxide:  $(C_4H_9)_3SnOH$ , diphenyltin (IV) oxide:  $(C_6H_5)_2SnO$  and triphenyltin (IV) hydroxide:  $(C_6H_5)_3SnOH$  were purchased from Sigma-Aldrich Chemical Company (Germany) and used as such. The solvents: methanol, propanol, and DMSO were Sigma- Aldrich products of analytical grade with purity ranging from 98-99.8 %.

### 2.1 Synthesis of potassium hydrogen ethanedioate (L)

Potassium hydroxide (0.05 mol, 2.8338 g) was completely dissolved in 50 mL distilled water in a 250 mL flat bottom flask containing a magnetic stirrer bar and ethanedioic acid (0.0241mol, 4.1920 g) was added and refluxed for 1 hour until all have reacted giving a clear solution. After cooling in an ice-bath, crystals of potassium hydrogen ethanedioate separated out and were filtered in a Buchner filtering unit and dried to a constant weight in a desiccator [12]-[15].

## 2.2 Synthesis of potassium dibutyltin (IV) ethanedioate $Bu_2SnL_2$ (1) and potassium diphenyltin (IV) ethanedioate: $Ph_2SnL_2$ (3)

Dibutyltin (IV) oxide, 1.9506 g (0.0080 mol) was refluxed in a methanol-n-propanol mixture of ratio 4:1 in a 250 mL flask for five (5) hours using Dean and Stark apparatus to give a clear solution of the intermediate: dibutyltin (IV) dipropoxide. Water in the solution distilled off as an azeotrope at 96-98 °C. After cooling, potassium hydrogen ethanedioate(L) 2.1063 g (0.008 mol) was added and refluxed for an hour and kept in an oven at 40 °C to obtain a white crystalline product (1) [12]-[19]. Similar procedure was used in the synthesis of  $Ph_2SnL_2$  (3) as in our earlier report [12],[14].

## 2.3 Synthesis of potassium tributyltin (IV) ethanedioate: $Bu_3SnL$ (2) and potassium triphenyltin (IV) ethanedioate: $Ph_3SnL$ (4)

Tributyltin (IV) hydroxide, 0.5000 g (0.0008 mol,) and potassium hydrogen ethanedioate, 0.2085 g (0.0008 mol) were suspended in methanol and refluxed for five hours between 60 °C to 70 °C in Dean and Stark apparatus. The methanol distilled off at 64.5 °C leaving a white precipitate which was kept in an oven for 72 hours at 40 °C to give white crystalline solid of  $Bu_3SnL$  (2). Same procedure [12]- [19] was also used in synthesizing  $Ph_3SnL$  (4)

## 2.4 Physicochemical Measurements

Fansworth and Pekola method [20] was adopted for tin content analysis. Melting points were obtained with Fisher-Johns microscope hot stage melting point apparatus and were not corrected. Fourier transform infrared spectra in the range 4000–400  $cm^{-1}$  were recorded using potassium bromide pellets on FTIR-8400S spectrophotometer (SHIMADZU).  $^1H$  and  $^{13}C$  NMR spectra were recorded at room temperature using NMR Nujol 400 MHz spectrophotometer.

## 2.5 Biological Investigations

The antibacterial activity of the synthesized compounds was tested on four Gram-positive bacteria: *Staphylococcus aureus*, *Streptococcus pyogenes*, *Bacillus cereus*, *Corynebacterium ulcerans* and four Gram-negative bacteria: *Escherichia coli*, *Klebsiella pneumoniae*, *Proteus mirabilis* and *Pseudomonas aeruginosa* in mueller hinton agar medium. The antifungal activity of the compounds was tested on the test fungi: *Microsporum audouinii*, *Microsporum distortum*, *Microsporum gallinae* and *Microsporum gypseum* in Sabouraud dextrose agar. Clinical isolates of the microbes used were obtained from Institute for Agricultural Research (I.A.R) as well as Veterinary Medicine and Medicinal Microbiological Department, Ahmadu Bello University Teaching Hospital, Zaria. Agar well diffusion technique and dilution method were used, as in our earlier report [12], [14].

### 2.5.1 Agar Well Diffusion Technique

The method adopted for determination of antimicrobial activity of the organotin compounds was agar well diffusion

technique. Sabouraud dextrose agar (SDA) and Mueller Hinton agar were used as culture media for fungi and bacteria respectively. They were prepared according to manufacturer's instructions, sterilized at 121 °C for 15 minutes, poured into sterile petri dishes under an aseptic hood and allowed to cool and solidify.

The sterile media were seeded with 0.1 mL of standard inoculums of the test microbes and spread evenly over the surfaces of the media using a sterile swab. A well was cut at the centre of each inoculated medium using a standard cork borer of 6 mm diameter and 200  $\mu g/mL$  of the test compounds dissolved in DMSO were introduced into their respective wells. Other wells supplemented with reference antifungal and antibacterial drugs: fluconazole & fulcin and erythromycin respectively were used as controls. The media were incubated at 30 °C for 7 days and at 37 °C for 24 hours for the fungi and bacteria respectively, and checked daily for inhibition zone: area where the microbes were unable to grow [21], [22]. Where inhibition zones were not observed, the organotin used was inactive or concentration used may be less than required.

### 2.5.2 Minimum Inhibition Concentration (MIC)- Broth Dilution Method

The minimum inhibition concentrations (MICs), the lowest concentrations of the test compounds exhibiting no visible growth of bacteria or fungi, were determined by agar dilution method. Stock solutions of the synthesized compounds (200  $\mu g/mL$ ) in DMSO were prepared, and serial dilution in sterile Mueller Hinton broth for determination of antibacterial activity and in Sabouraud dextrose broth medium for antifungal activity was made to obtain the concentrations of 100  $\mu g/mL$ , 50  $\mu g/mL$ , 25  $\mu g/mL$  and 12.5  $\mu g/mL$ . The broth were prepared in test tubes, sterilized at 121 °C for 15 minutes and allowed to cool.  $1.5 \times 10^5$  CFU/mL of test fungi and  $1.5 \times 10^8$  CFU/mL of test bacteria in normal saline were made and introduced separately into each of the concentrations and incubated at 30 °C for 7 days (fungi) and at 37 °C for 24 hrs (bacteria) [20]. The test tubes were observed for turbidity (growth) and the lowest concentration of a compound in the broth which showed no turbidity was recorded as the MIC. Contents of MIC in the serial dilution were sub cultured onto the prepared medium and incubated at 30 °C for 7 days and 37°C for 24 hrs for fungi and bacteria respectively. Plates were observed for colony growth, MFC was the plate with lowest concentration of compound without colony growth [4], [12]-[14]. Minimum fungicidal concentration (MFC) was determined in order to ascertain if the test microbes were completely killed or only inhibited.

## 3. Results and Discussion

### 3.1 Synthesis

Synthesis of organotin (IV) derivatives of potassium hydrogen ethanedioate (1, 2, 3 and 4) was successfully achieved from their oxides [ $(C_4H_9)_2SnO$ ,  $(C_6H_5)_2SnO$ ] and hydroxides [ $(C_4H_9)_3SnOH$ ,  $(C_6H_5)_3SnOH$ ], respectively. The reactions occurred in eight steps as shown in scheme 1. The ligand: HOCCOOK was first prepared by the reaction

between KOH and ethanedioic acid: HOOC-COOH according to route (I).  $(C_4H_9)_2SnO$  and  $(C_6H_5)_2SnO$  were refluxed separately in 4:1  $CH_3OH$  and  $C_3H_7OH$  (route II) yielding their respective propoxides as intermediates. These were further reacted with the ligand L HOOC-COOK to produce compounds 1 and 3 (routes III and IV).  $(C_4H_9)_3SnOH$  and  $(C_6H_5)_3SnOH$  were refluxed in  $CH_3OH$  which gave their

dimethoxides as intermediates (routes V and VI respectively) which were further reacted with the ligand: HOOC-COOK to produce compounds 2 and 4 (VII and VIII). Water produced in the process was collected in the separator of Dean and Stark apparatus and removed from the reaction.

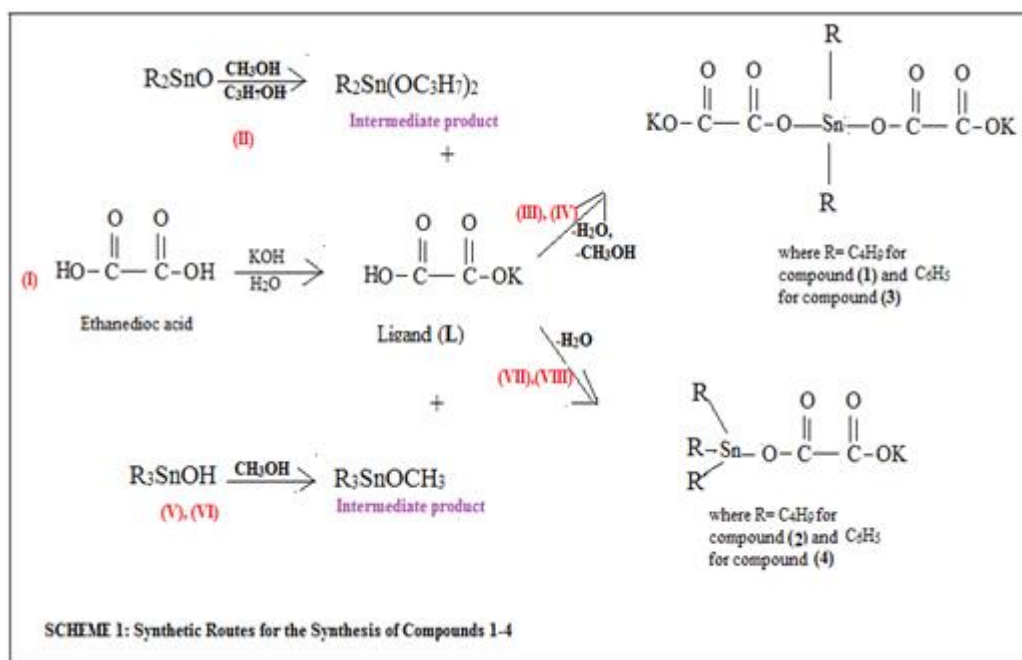


Table 1, shows the physicochemical data of synthesized compounds. Observed % Sn was found to be in agreement with calculated tin content, an indication that the products were formed. The melting points of the compounds reflect their purity.

**Table 1:** Physicochemical Data of compounds synthesized

Compound no.	%Sn	Mpt.	%yield	
1*	24.43 <sup>a</sup>	24.49 <sup>b</sup>	340 – 344	85.79
2*	28.46	28.54	402 – 405	92.48
3*	21.87	21.58	288 – 289	95.68
4*	24.91	24.94	300-301	92.16

a=found, b=theoretical, \* = white crystals

### 3.2 Infrared Spectroscopy

Important Infrared absorption bands of synthesized compounds are listed in Table 2.

**Table 2:** Important IR bands of synthesized compounds( $cm^{-1}$ )

Compound no.	1	2	3	4	L
$\nu_{asym}(COO)$	1629	1643	1677	1663	1694
$\nu_{sym}(COO)$	1460	1464	1423	1432	1442
$\Delta\nu$	169	179	254	230	252
Sn-O	411	501	515	445	-
Sn-O-C	1017	1029	906	906	-
Sn-Bu	671	719	-	-	-
Sn-Ph	-	-	1083	1072	-
C-H <sub>arom</sub>	-	-	3050	3058	-
O-H	-	-	-	-	3486

L = HOOC-COOK

The Fourier transform infrared spectrum of the ligand(L) showed characteristic stretching absorption bands at 3486

$cm^{-1}$ , 1694  $cm^{-1}$  and 1442  $cm^{-1}$ , which were assigned to  $\nu(OH)$ ,  $\nu(COO)_{asym}$  and  $\nu(COO)_{sym}$ , respectively. The spectra of compounds (1-4) revealed absence of a strong vibrational frequency due to OH stretching at 3486  $cm^{-1}$  which was present in the ligand. The presence of bands in the range 515-411  $cm^{-1}$  and 1029-906  $cm^{-1}$  indicated deprotonation of -COOH group and formation of new Sn-O and Sn-O-C bonds, respectively [23], [24]. Formation of the compounds was further confirmed by appearance of medium intensity bands in the range 1083-671  $cm^{-1}$  due to Sn-C-O, Sn-Bu and Sn-Ph bonds. The band at 3486  $cm^{-1}$  which appeared in the free ligand as the  $\nu(O-H)$  stretching vibrations but absent in compounds 1-4, indicated metal-ligand bond formation through these sites [25]. The red shifts of the bands with respect to the free acid also served to confirm the formation of organotin carboxylates [10]. The binding mode of ligand to tin atom was determined by the difference between the asymmetric and symmetric carboxylate stretching vibrations,  $\Delta\nu = \nu_{asym}(COO)$  and  $\nu_{sym}(COO)$  [26]. Generally, it is believed that  $\Delta\nu$  value  $< 200$   $cm^{-1}$  indicates that the carboxylate moiety is bidentate, while  $>200$   $cm^{-1}$  indicates monodentate [5],[23], [25]. The magnitude of  $\Delta\nu$  of 179 – 169  $cm^{-1}$  for complexes 1 and 2 indicated that the carboxylate ligands function as bidentate under the conditions employed while that of compounds 3 and 4 as well as the ligand (L) with values in the range 254 – 230  $cm^{-1}$  indicated their carboxylate groups as monodentate. It is, therefore, proposed that the carboxylate group in these compounds are acting as both bidentate and monodentate ligand [10], [27], [28].

### 3.3 NMR Spectroscopy

The  $^1\text{H}$  NMR spectra of synthesized compounds are shown in Table 3. Signals of the protons in all the compounds were observed within the expected range. The expected aliphatic and aromatic peaks with correct integration and multiplicities were observed. Compounds **3** and **4** showed complex pattern in the range 7.34 - 7.76 ppm due to the aromatic protons of phenyl groups. Figure 1, shows the numbering of protons and carbons in the structure of the compounds.

**Table 3:**  $^1\text{H}$  NMR data of synthesized compounds and ligand

Compound no.	1	2	3	4	L
a	1.23	-	-	-	-
b	1.41m	-	-	-	-
c	0.82t(7.2)-	-	-	-	-
g	1.02	-	-	-	-
$\beta$	-	-	7.76	7.66	-
$\gamma$	-	-	7.34s	7.37	-
$\delta$	-	-	7.51	7.47	-
O-H	-	-	-	-	9.99

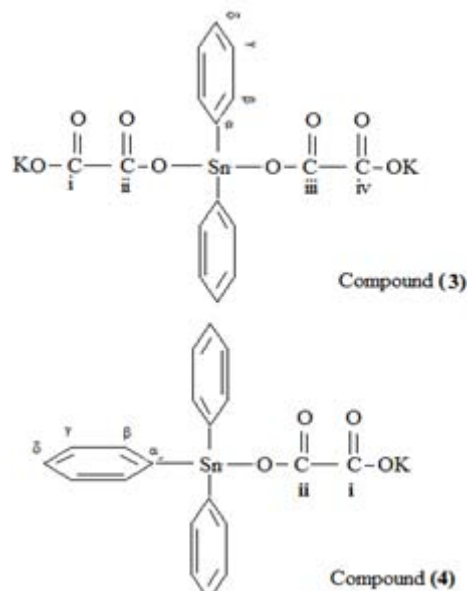
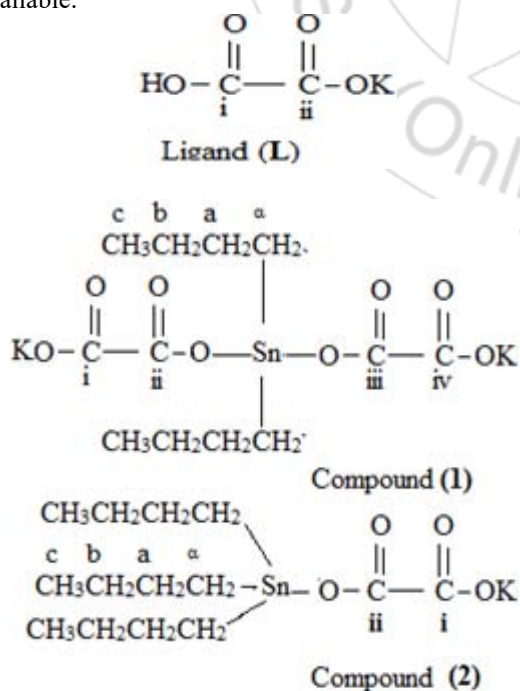
L = HOCCOOK

a) Chemical shift( $\delta$ ) in ppm, J( $^1\text{H}$ - $^1\text{H}$ )

b) Multiplicity is given as s = singlet, t = triplet, m = multiplet

c) Figure 1 shows the numbers assigned to protons and carbons in the proposed structure of synthesized compounds for ease of reference.

Butyl protons found in compounds **1** designated **a** and **b** in fig. 1, showed complex peaks due to  $-\text{CH}_2-\text{CH}_2-\text{CH}_2-$  in the range 1.23 - 1.41 ppm and a clear triplet due to the terminal methyl group designated **c** around 0.82 ppm with ( $^1\text{H}$ - $^1\text{H}$ ) coupling of 7.2 Hz. The methylene protons designated **a**, showed peak at 1.02 ppm. The  $^1\text{H}$  NMR for compound **2** is not available.



**Figure 1:** Numbering of protons and carbons in the Structure of synthesized compounds (1-4)

$^{13}\text{C}$  NMR spectral data of compounds **1-4** are given in Table 4. The carboxyl carbons (**i**, **ii**, **iii** and **iv**) of all compounds were assigned signals in the range 162.64 - 175.74 ppm which are in agreement with literature [18]. The C=O resonance group of the complexes at  $\delta$  162.64 - 175.74 ppm were shifted down from the position in the free ligand, which appeared at  $\delta$  188.17 ppm. The shift could be due to a decrease in electron density at the carbon atoms when oxygen bonded to metal ion [15], [29]. This observation indicates that, complexation in the compounds occurred through the oxygen atoms of the carboxylate group [18], [28].

**Table 4:**  $^{13}\text{C}$  NMR data of synthesized compounds and ligand (ppm)

Compound no.	1	2	3	4	L
i,iv	162.64	167.58	172.01	175.74	188.17
ii,iii	162.64	167.58	172.01	175.74	188.17
a	39.75	37.34	-	-	-
b	29.78	26.69	-	-	-
c	28.87	28.28	-	-	-
g	26.49	25.98	144.54	137.36	-
$\beta$	-	-	136.83	128.08	-
$\gamma$	-	-	129.04	127.93	-
$\delta$	-	-	137.05	135.82	-

L = HOCCOOK

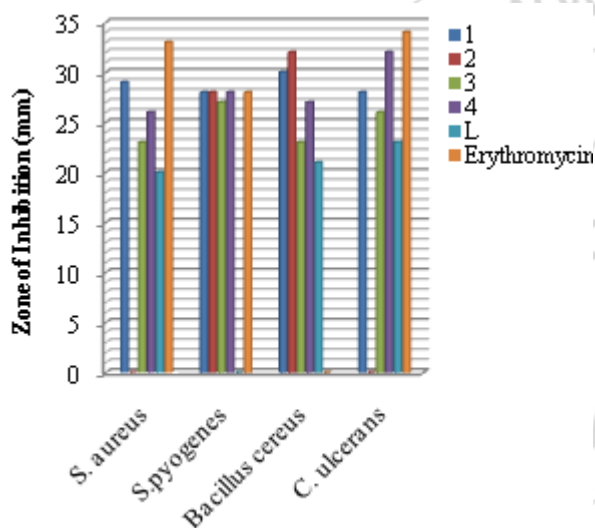
Signals for butyl carbons (**a** and **b**) appeared at 26.69 - 39.75 ppm while  $-\text{CH}_3$  designated **a**, appeared at 26.49 ppm and 25.98 ppm, respectively for compounds **1** and **2**. Phenyl carbons were assigned signals in the range 128.03 - 144.54 ppm with  $\alpha(\text{Sn}-\text{C})$  at 144.54 ppm and 137.36 ppm for compounds **3** and **4**, respectively. These signals are in agreement with our earlier report [13], [14].

### 3.4 Biological Activity

#### 3.4.1 Antibacterial Activity

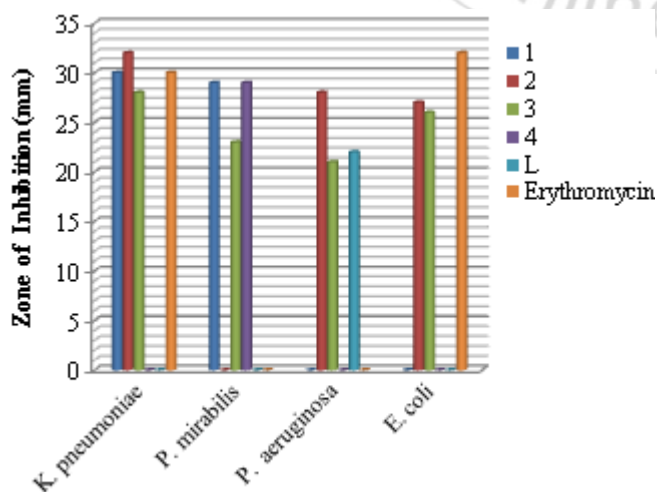
Antibacterial activity tests of the ligand (L) and its organo-tin (IV) compounds (**1-4**) were carried out against eight bacterial

strains; four Gram-Positive (*Staphylococcus aureus*, *streptococcus pyogenes*, *Bacillus cereus* and *corynebacterium ulcerans*) and four Gram-negative bacteria (*Escherichia coli*, *Klebsiella pneumoniae*, *Proteus mirabilis* and *Pseudomonas aeruginosa*). The results are shown in fig. 2 and 3, respectively. Erythromycin was used as standard drug in this assay. Reports have shown that criteria for activity is based on zone of inhibition (mm); inhibition zone more than 20 mm shows significant activity, for 18-20 mm inhibition activity is good, 15-17 mm is low, and below 11-14 mm is non-significant [23],[24],[27]. The results revealed that all the synthesized compounds showed significant activity against all tested bacterial strains with zones of inhibition ranging from 21-32 mm. However, there were few cases where the compounds, ligand and standard drug did not show activity against some strains. Generally, compounds 1-4 were more active against gram-positive bacteria than the gram-negative ones. Even though, erythromycin showed higher activity ranging from 28-34 mm, it showed no activity against *Bacillus cereus*, *Proteus mirabilis* and *Pseudomonas aeruginosa*.



Gram-positive bacteria

Figure 2: Zone of Inhibition of compounds against gram positive bacteria



Gram-negative bacteria

Figure 3: Zone of Inhibition of compounds, drug and ligand against gram positive bacteria

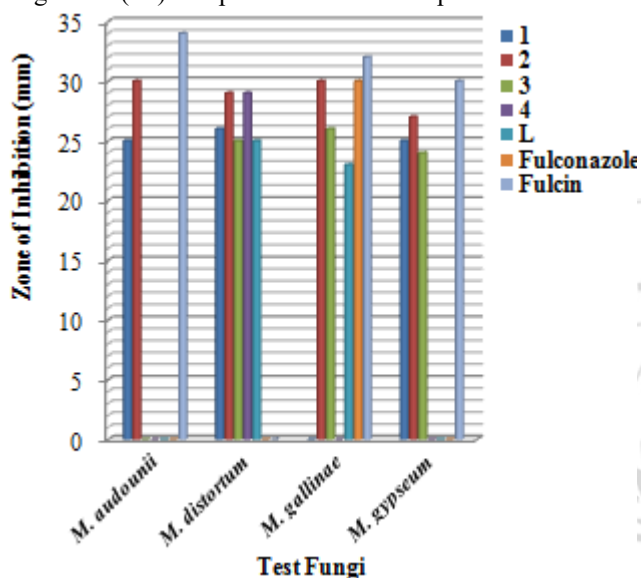
It was noted that compounds 1, 3 and 4 exhibited significant activity against *Staphylococcus aureus* (29, 28 and 26 mm), *streptococcus pyogenes* (28,27 & 28mm), *Bacillus cereus* (30, 23 & 27 mm) and *corynebacterium ulcerans* (28, 26 & 32 mm) with compound 4 exhibiting the highest activity against *corynebacterium ulcerans* (32 mm) very close to the activity of erythromycin. The ligand L, showed no activity against *streptococcus pyogenes* but exhibited lowest activity for the other gram-positive bacterial strains. Despite the significant activity of the standard drug used, it did not show any activity against *Bacillus cereus* but compounds 1-4 did. Compounds 1, 2 and 3 exhibited the same activity with erythromycin (28 mm) against *streptococcus pyogenes* (fig. 2). For the gram-negative bacteria, Fig. 3 demonstrates that compounds 1 and 2 showed significant activity against *Klebsiella pneumoniae* and also greater than the reference drug while compound 3 exhibited significant activity (28 mm) very close to the standard drug. On the other hand, compound 4 and L exhibited no activity against the said bacteria. Compound 4 and L only exhibited activity against *Proteus mirabilis* (20 mm) and *Pseudomonas aeruginosa* (28 mm) respectively. The antibacterial study also demonstrates that compound 3 exhibited significant activity against all the gram-negative bacteria (*Escherichia coli*, *Klebsiella pneumoniae*, *Proteus mirabilis* and *Pseudomonas aeruginosa*). In general, compounds 2 and 4, the triorganotin (IV) compounds showed greater antibacterial activity than compounds 1 and 3, the diorganotin (IV) counter parts. This is consistent with literature and may be due to greater lipophilicity and permeability through the cell membrane [23], [24], [27]. MIC of the compounds was obtained at the concentrations of 15 µg/mL while their MFC revealed that all the bacteria were not just inhibited but were completely killed at the same concentration.

### 3.4.2 Antifungal Activity

The ligand L and its organotin (IV) compounds (1-4) were screened for antifungal activity against four fungal strains (*Microsporium audouinii*, *Microsporium distortum*, *Microsporium gallinae* and *Microsporium gypseum*) by using agar tube dilution method [21] – [23]. From the result shown in fig. 4, it was observed that all the compounds exhibited significant activity against test fungi with compounds 2 and 4 (triorganotin (IV) compounds) exhibiting the highest activity against *Microsporium audouinii* & *Microsporium gallinae* (29 mm) and *Microsporium distortum* (28 mm), respectively. These activities were close to those exhibited by the standard drugs fulcin and fluconazole (30-34 mm). Despite the significant activity exhibited by both standard drugs, they did not exhibit activity against *Microsporium distortum* but compounds 1-4 showed significant activity with zones of inhibition ranging from 24 – 28 mm. Fulcin, only exhibited activity against *Microsporium gallinae* (30 mm). Of all the compounds synthesized, only compound 2 exhibited activity against all the test fungi with zones of inhibition ranging from 26 - 29 mm. Compound 4 showed activity only against *Microsporium distortum* at the concentrations used. Overall, compounds 1 – 4 showed good antifungal activity at the concentration used. MIC of the compounds was obtained at the concentrations of 25 µg/mL while their MFC revealed that all the fungi were not just inhibited but were completely killed at the same concentration.

The increased antibacterial and antifungal activities observed in compounds **1- 4** than their ligand could be due to the presence of metal ions introduced into their structures when the ethanedioic acid (HOOC<sub>2</sub>COOH) was coordinated with K<sup>+</sup> and Sn<sup>4+</sup>. This agrees with the known fact that many biologically active compounds become more active upon complexation than in their uncomplexed forms [18]. Findings have shown that organotin (IV) carboxylates are more active than their ligand which is in agreement with literature [15, 18].

The biological activity of organotin compounds especially diorganotin (IV) compounds have been reported



**Figure 4:** Zone of Inhibition of compounds, drug and ligand against test fungi

to depend solely on the organotin moiety; R<sub>2</sub>Sn<sup>2+</sup> and R<sub>3</sub>Sn<sup>+</sup> [5]. Carboxylate groups are known to influence the delivery of organotin (IV) moiety to the point of action. Therefore, the activities of compounds **1- 4** appeared to be a combined effect of the metal ions and carboxylate groups. Since diorganotin (IV) compounds (**1** and **3**) are not known for their high biological activities, their activity in this study could probably be due to the presence of potassium and Sn ions in their structures [29].

#### 4. Conclusion

A series of four organotin (IV) carboxylate compounds of potassium hydrogen ethanedioate synthesized and characterized by FT-IR, <sup>1</sup>H NMR and <sup>13</sup>C NMR, FTIR spectra revealed that the modes of coordination in compounds (**1**) and (**2**) are bidentate and in compounds (**3**) and (**4**) are monodentate while NMR confirmed the formation of compounds. Generally, all the compounds exhibited significant antimicrobial activity comparable to standard drugs used. Compound (**4**), Ph<sub>3</sub>SnL exhibited antibacterial activity more than its diphenyl counterpart, compound (**3**): Ph<sub>2</sub>SnL<sub>2</sub> while compound (**1**), Bu<sub>2</sub>SnL<sub>2</sub> exhibited higher activity than compound (**2**), Bu<sub>3</sub>SnL against gram-positive bacteria. The observation that the triorganotin (IV) compounds :(**4**) is more active agrees with the notion that the number of carbon atoms in an organotin moiety affects biological activity [29] of the compound. For gram-

negative bacteria, compound (**2**), Bu<sub>3</sub>SnL exhibited more activity than compound (**1**), Bu<sub>2</sub>SnL<sub>2</sub> while compound (**3**) Ph<sub>2</sub>SnL<sub>2</sub> exhibited more activity than compound (**4**), Ph<sub>3</sub>SnL. Antifungal activity revealed that Bu<sub>3</sub>SnL exhibited activity more than its Bu<sub>2</sub>SnL<sub>2</sub> counterpart while Ph<sub>3</sub>SnL was more active than Ph<sub>2</sub>SnL<sub>2</sub> its counterpart. The order for the antifungal activity in this study was Bu<sub>3</sub>SnL > Ph<sub>3</sub>SnL > Ph<sub>2</sub>SnL<sub>2</sub> > Bu<sub>2</sub>SnL<sub>2</sub>. The order of activity against gram-positive bacteria was Ph<sub>3</sub>SnL > Bu<sub>2</sub>SnL<sub>2</sub> > Ph<sub>2</sub>SnL<sub>2</sub> > Bu<sub>3</sub>SnL. The order of activity against gram-negative bacteria was Bu<sub>3</sub>SnL > Bu<sub>2</sub>SnL<sub>2</sub> > Ph<sub>2</sub>SnL<sub>2</sub> > Ph<sub>3</sub>SnL.

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